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(54) Title: RESIN AND COMPOSITE WOOD PANELS

(57) Abstract

A binder composition comprising a formaldehyde-based resin and an isocyanate containing compound wherein said formaldehyde-based resin comprises formaldehyde present in a molar ratio of formaldehyde to comonomer in the range of from about 0.2:1 to about 5:1 and wherein the isocyanate containing compound is present in an amount in the range of from about 0.5 to about 50 % by weight of the binder composition.

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# RESIN AND COMPOSITE WOOD PANELS

The present invention relates to resin for use in the manufacture of composite panels, composite panels and method of manufacture therefore. In particular, the present invention relates to formaldehyde based resin for use in the manufacture of composite board, which composite board has low formaldehyde emissions. Desirably, formaldehyde emissions are reduced to a level approaching the formaldehyde emission of timber.

Wood composite panel products such as particle board and medium density fibre board ("MDF") are generally manufactured using urea formaldehyde ("UF") resins. In the past UF resins have been manufactured with an excess of formaldehyde to impart increased cure speed and bond strength of the cured resin system. There are two principle disadvantages associated with the use of UF resin. These are a lack of resistance to weather and water, and a susceptibility to emission of formaldehyde vapours.

The lack of weather resistance may be tolerated in certain applications, such as where composite boards are intended for interior use. However, the tendency of UF resins to emit formaldehyde is a serious disadvantage that severely effects their use in interior applications. The emission of formaldehyde has been a long term problem in the industry and intensive work in the area of reducing emissions began in earnest in the 1970's. Confronted with the introduction in many countries of stringent formaldehyde emission requirements (for example Japan and Europe's E1 and E0 consumer recommendations), resin manufacturers have developed UF resins with decreased formaldehyde emissions. This has generally been achieved by producing resin with a lower formaldehyde: urea (F:U) mole ratio. Historically, the F:U ratio used was of the order of 1.4:1. More recently, mole ratios of 0.85:1 have been employed to achieve reduced emissions. However, decreasing the F:U mole ratio results in the UF resin becoming more difficult to handle, bond strength decreases and consequently higher resin contents have been found to be necessary in the composite panels, together with significantly longer pressing times. Low formaldehyde emitting UF resin formulations have compromised strength and manufacturing costs for emission low formaldehyde.

There is increasing pressure from wood composite panel manufacturers and government bodies to further reduce the level of formaldehyde emissions of composite panel to levels similar to those of natural wood, the so-called "E0" level. Manufacturers wanting to achieve composite panel with E0 emissions have had limited alternatives. Formaldehyde free binders such as 4, 4'- methylenebis (phenyl isocyanate) ("MDI") may be used. However, while the use of MDI as a unitary binder avoids the problem of formaldehyde emissions, it is relatively expensive. UF resins with low formaldehyde content which have been modified with melamine or phenol may also be used. However, the use of these modified UF resins may result in composite panels with improved dimensional stability, panels may still exhibit unacceptable levels of formaldehyde emissions.

There exists a need for a resin for use in the manufacture of composite panel which addresses the problem of formaldehyde emission and/or lack of dimensional stability, or provides a useful alternative to composite panel products currently available.

In one aspect, the present invention provides a binder composition comprising a formaldehyde-based resin and an isocyanate containing compound wherein said formaldehyde-based resin comprises formaldehyde present in a molar ratio of formaldehyde to comonomer in the range of from about 0.2:1 to about 5:1 and wherein the isocyanate containing compound is present in an amount in the range of from about 0.5 to about 50% by weight of the binder composition.

The binder composition may be supplied to a particle board manufacturer as a two-component system, the formaldehyde-based resin will generally be supplied separately to the isocyanate containing compound. Such a two component system represents a further aspect of the invention. Other additives may also be supplied separately or blended with either the formaldehyde-based resin or the isocyanate containing compound.

In another aspect, the present invention provides a composite board comprising cellulose-based particles and a binder composition comprising a formaldehyde-based resin and an isocyanate containing compound wherein said formaldehyde-based resin comprises formaldehyde present in a molar ratio of formaldehyde to comonomer in the range of from about 0.2:1 to about 5:1 and wherein the isocyanate containing compound is present in an amount in the range of from

about 0.5 to about 50% by weight of the binder composition.

In another aspect the present invention provides a method of manufacturing a composite panel comprising the steps of :

(a) blending cellulose-based particles with a binder composition comprising a formaldehyde-based resin and an isocyanate containing compound wherein said formaldehyde-based resin comprises formaldehyde present in a molar ratio of formaldehyde to comonomer in the range of from about 0.2:1 to about 5:1 and wherein the isocyanate containing compound is present in an amount in the range of from about 1 to about 50% by weight of the binder composition; and

(b) curing the binder composition to form a composite panel.

A composite panel manufactured according to this process also represents a further aspect of the invention.

The binder composition comprises a formaldehyde-based resin. Formaldehyde-based resins incorporate formaldehyde along with a comonomer or comonomers. For convenience, throughout this specification and claims which follow, the term "comonomer" will be used to refer to one or more comonomers suitable for forming a formaldehyde-based resin.

The molar ratio of formaldehyde to comonomer is, in binary systems, such as urea-formaldehyde systems, the molar ratio of formaldehyde to comonomer is simply the molar ratio of the formaldehyde to the comonomer (i.e. urea). In the ternary, or more complex systems, the molar ratio of formaldehyde to comonomer is the molar ratio of the formaldehyde to the sum of the molar amounts of the comonomers.

The formaldehyde-based resin comprises formaldehyde present in a molar ratio of formaldehyde to comonomer is in the range of from about 0.2:1 to about 5:1. Preferably the molar ratio of formaldehyde to comonomer is in the range of about 0.3:1 to about 4:1. More preferably the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 3.5:1.

Suitable comonomers for use in the formaldehyde-based resin include polyfunctional amines, phenols, and other comonomers capable of forming copolymers with the formaldehyde.

The polyfunctional amines comprise two or more primary, secondary and or tertiary amine groups. Examples of suitable polyfunctional amines include melamine, urea, guanidines, para-toluene sulfonamide, triazines, thiourea and dicyandiamide. Preferred polyfunctional amines include melamine and urea.

Preferably the polyfunctional amine is urea. Examples of suitable phenols include phenol, resorcinol, tannins, lignins, bisphenol A, cresol and xylenol.

Preferably, the formaldehyde-based resin may be selected from the group consisting of urea formaldehyde resins, phenol formaldehyde resins, phenol urea formaldehyde resins, melamine urea formaldehyde resins, phenol melamine urea formaldehyde resins, phenol melamine formaldehyde resins.

In urea formaldehyde resins it is preferable that the molar ratio of formaldehyde to comonomer is in the range of from about 0.3:1 to about 1.5:1, more preferably from about 0.4:1 to about 1.1:1, even more preferably about 0.4:1 to about 0.9:1, and most preferably about 0.45 to about 0.75.

In these urea formaldehyde resins part of the urea may be replaced with melamine. The modification of urea formaldehyde resins with melamine may provide improved water resistance to the binder composition, as well as the composite panels resulting in improved dimensional stability.

The melamine may replace up to 85% by weight of comonomer. The urea formaldehyde resin modified with melamine may include a melamine component of from about 0.5 to about 60% and preferably from 1 to 50% weight on solids.

urea formaldehyde resins are preferably formulated to a viscosity of up to about 700 cps and more preferably in the range of about 30 to 600 cps. Preferably, when it is desired to use the urea formaldehyde resins in applications where the stability of the unpressed composite is an important factor, the viscosity is in range of from about 300 to about 500 cps. It is also possible to use the urea formaldehyde having a viscosity is in the range of from about 30 to about 60 cps.

Preferably, the urea formaldehyde resin comprises from about 40% to about 70% by weight solids, more preferably from about 35% to about 70% by weight solids.

Preferably, the urea formaldehyde resin has a pH in the range of from about 8 to about 10, more preferably from about 8.5 to about 9.5.

In phenol formaldehyde resins it is possible to formulate the resin at either acid curing or alkaline curing conditions. Under acid curing conditions it is preferable to use a high phenol content. Preferably the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 1:1, more preferably from about 0.4:1 to about 0.9:1, and most preferably about 0.45:1 to about 0.7:1.

Under alkaline curing conditions it is preferable to use a low phenol content. Preferably the molar ratio of formaldehyde to comonomer is in the range of from about 1.8:1 to about 4:1, more preferably from about 2:1 to about 2.5:1, and about 3.5:1 to about 3.8:1.

In these phenol formaldehyde resins part of the phenol may be replaced with urea.

The urea may replace up to 50% weight on solids. Preferably the urea may be present in an amount of from about 1% to about 25% weight on solids.

PF resins are preferably formulated to a viscosity of up to about 400 cps and more preferably in the range of about 30 to 400 cps. Preferably, when it is desired to use the phenol formaldehyde resins in applications where the stability of the unpressed composite is an important factor, the viscosity is in range of from about 200 to about 400 cps. It is also possible

to use the phenol formaldehyde resins having a viscosity is in the range of from about 30 to about 60 cps.

Preferably, the phenol formaldehyde resin comprises from about 30% to about 50% by weight solids, more preferably from about 35% to about 45% by weight solids.

Preferably, the phenol formaldehyde resin has a pH in the range of from about 7 to about 12, more preferably from about 9 to about 12, such as from about 10 to about 12.

A variety of additives such as those used in the manufacture of conventional formaldehyde resins may incorporated into the formulations of the formaldehyde based resin of the present invention. pH modifying agents such as acids, bases and buffers. Acids typically used may include formic acid, hydrochloric acid and sulfuric acid. Bases typically used may include sodium hydroxide and potassium hydroxide. Buffers which may be used include triethanolamines and borax. Other additives include hexamine, which advantageously breaks down to formaldehyde *in situ*, sulfites, polyvinyl alcohol and sodium metabisulfite.

We have found that the formaldehyde-based resin component may advantageously be blended with a release agent for supply to a fabricator of composite boards. When using isocyanate based binders, fabricators will generally employ release agents to facilitate removal of the boards from the presses. Generally release agents such as waxes are independently applied to the cellulose fibres. The incorporation of release agents into the formaldehyde-based resin allows greater convenience in the manufacturing process, the fabricator may operate a reduced stock inventory and reduces likelihood of incorrect dosages being used. Alternatively the release agent may be included in the isocyanate component of the system.

Preferably, suitable release agents include paraffin and synthetic waxes such as montan waxes, polyethylene waxes and polypropylene waxes. Generally the release agent may be present in the formaldehyde based resin an amount of at least 5% by weight solids of the isocyanate based compound, preferably at an amount of about 10% by weight solids of the isocyanate based compound.

Release agents may preferably be incorporated into the formaldehyde-based resin in the form of an emulsion. An emulsion of the release agent may be formed by emulsifying a blend of the release agent and water in the presence of a surfactant. Preferably the blend is heated to an elevated temperature which promotes the formation of the emulsion. Suitable surfactants for promoting the formation and stabilisation of the emulsion include ethylene oxide derivatives.

Alternatively, the release agent may be incorporated in an unemulsified form. This advantageously enables the release agent to be incorporated without the need for additional surfactant.

The release agents may preferably be incorporated into the formaldehyde resin in the final stages of manufacture or may be incorporated by blending with the formaldehyde resin after the manufacture of the formaldehyde resin has been completed. Typically, the release agent may be added as an emulsion. Alternatively the release agent may be added in solid form.

Preferably, the isocyanate containing compound may be a polyisocyanate including blocked isocyanates. Suitable polyisocyanates include toluene diisocyanate ("TDI") and 4, 4'-methylenebis (phenyl isocyanate). Preferably the isocyanate containing compound for use in the present invention is 4,4'-methylenebis (phenyl isocyanate) or commercial alternatives, generally referred to in the art as MDI. One example of a suitable MDI is Rubinate DUO B200 (ex Orica Australia Pty Ltd). The isocyanate containing compound may be provided in a variety of forms, such as neat or in an emulsified form.

The isocyanate containing compound for use in the present invention is present in the binder composition in an amount in the range of from 0.5 to 50% by weight of said binder composition. Preferably the isocyanate containing compound is present in the binder composition in an amount in the range of from 5 to 30%, more preferably from 10 to 25%.

In one aspect the formaldehyde-based resin is preferably pre-reacted with up to 50% w/w on resin solids of melamine. The binder composition manufactured from such a pre-reacted

formaldehyde-based resin may impart superior cold and hot water swell properties to the composite board, i.e. improved dimensional stability.

The cellulose-based particles selected for use in the manufacture of composite panels will be dependant upon the nature of the composite panels to be produced. The cellulose-based fibres, particles or chips may be in the form of raw fibres. The cellulose-based fibres, particles or chips may be in the form of fibres which have not been pre-dried. In a preferred method the cellulose-based fibres, particles or chips may be in the form of untreated, moist raw fibres. Composite panels which may be manufactured in accordance with the present invention include particle board, medium density fibreboard, oriented strand board (OSB) plywood and combinations thereof fibreboard. such as tri-board which is a combination of oriented strand board and medium density fibreboard.

Particle board may be made from small discreet particles of wood. The wood particles may be made by cutting or breaking of the wood, their shape not being narrowly critical to the construction of the particle board. The particles of wood generally contain a moisture content of from 2 to 10% by weight. The manufacture of particle board generally combines a mechanical mixing of the particles and the binder composition followed by the application of heat and pressure so as to cure the resin and form the particle board. Typically the curing temperatures are in the range of from 130°C to 240°C although, dependent on the formulation other temperatures may be possible. Generally particle boards contain from 3 to 40% by weight of resin, preferably from 5 to 20% by weight of resin.

In a particularly preferred embodiment of the present invention the composite board is a medium density fibreboard. The medium density fibreboard comprises cellulose-based fibres which are in the form of a wood pulp. The binder is added to the wood pulp and the mixture dried to form a mat of dried fibres and binder. The temperatures chosen for the drying of the mixture are preferably such that the mixture is dried whilst the binder is not subject to conditions which will induce substantial curing. The dried mat is consolidated into the desired preform which is subsequently subjected to heat and pressure so as to cure the binder and produce the desired composite board. The cellulose fibre may be dried at temperatures ranging from 80°C to 140°C.

In the method of the present invention the cellulose-based particles may be blended with the formaldehyde-based resin, either where the formaldehyde based resin is premixed with the isocyanate containing compound or where the components are simply added simultaneously. Alternatively one or more components of the binder composition may be pre-blended with the cellulose-based particles and the remaining components subsequently combined with the pre-blended materials. For example, the cellulose-based particles may be blended with the formaldehyde-based resin and subsequently the isocyanate containing compound may be incorporated into the pre-blended composition. Alternatively, a formaldehyde-based resin and an isocyanate containing compound may be blended with the cellulose particles and subsequently further comonomer may be added to the pre-blended components to decrease the molar ratio of formaldehyde to comonomer.

The blending of the cellulose particles with the resin composition may be performed *in situ*, such as in a blender or a blow line.

Generally the curing of the binder composition to form a composite board may be by the process of hot pressing where the curing of the binder composition is effected at elevated temperatures and under applied pressure.

Typically the binder composition will be present in the composite board in the range of from 3 to 40% by weight. The addition rate of the binder composition will generally be dependant upon the physical properties which are required for the composite board and will be dependant upon the nature of the cellulose particles. Preferably the binder composition is present in the composite board in the range of from 5 to 30% by weight.

Panel products for use in non-humid applications where low formaldehyde emissions are required, a urea formaldehyde based binder with a low formaldehyde to urea ratio can be employed in conjunction with an isocyanate compound. This technique will provide an end product with surprisingly low emissions and unexpectedly superior excellent physical properties.

Panel products for use in humid conditions or where higher dimensional stability is required, MUF or more durable formaldehyde base resins such as phenol formaldehyde can be employed in conjunction with an isocyanate compound. These panel products will have high dimensional stability and low levels of formaldehyde emission. In order to further reduce formaldehyde emissions lower formaldehyde to comonomer ratios can also be employed. In order to improve general performance, binder loadings may be increased to achieve the desired physical properties.

Advantageously, there may be no need for release agents which are typically used with, for example, MDI. The binder composition may be reacted before the fibre is dried and pressed. However, in the present invention, the reaction may be generally slower and may not be complete before the fibre may be dried and pressed. This is particularly advantageous in obtaining a composite board having acceptable structural properties and dimensional stability.

Throughout this specification and the claims which follow, unless the context requires otherwise, the word "comprise", and variations such as "comprises" and "comprising", will be understood to imply the inclusion of a stated integer or step or group of integers or steps but not the exclusion of any other integer or step or group of integers or steps.

The present invention will now be described with reference to the following non-limiting examples.

## EXAMPLES

### EXAMPLES 1 and 2

1. Hybrid binders

   In Example 1, 100 solid parts of Sylvic DUO A110 (ex Orica Australia Pty Ltd), a urea formaldehyde resin, was mixed with 8.3 parts of Rubinate DUO B200 (ex Orica Australia Pty Ltd), an MDI, and 1.28 solid parts of montan wax to form the binder composition. In Example 2, 12.5 parts of Rubinate DUO B200 was used.

2. Manufacture of MDF

The binder compositions described above were applied to wood fibres in an MDF process via a blowline. In Example 1, the binder composition was applied at a rate of 13.1 solid parts of binder to 100 parts of dry wood. In Example 2, the binder composition was applied at a rate of 9.10 solid parts of binder to 100 parts of dry wood. The wood fibre was then pressed into MDF panels. The panels of Example 1 were pressed and cured at a temperature of 145°C. The panels of Example 2 were pressed and cured at a temperature of 180°C.

The resultant panel results are as follows:

| Example | Panel Thickness (mm) | Density (kg/m$^3$) | Formaldehyde emission* (mg/100g dry board) | IB (kPa) | MOR (MPa) | 24 hour swell (%) |
|---|---|---|---|---|---|---|
| 1 | 3 | 859 | 5.5 | 1300 | - | 19.3 |
| 2 | 15 | 741 | 4 | 810 | 45.8 | 5.8 |

* Measured using European Standard EN 717-3 1996 using a 24 hour incubation period.

**EXAMPLES 3 and 4**

1. Hybrid binder

100 solid parts of Sylvic DUO A120 (ex Orica Australia Pty Ltd), a MUF resin, was mixed with 27.3 parts of Rubinate DUO B200 (ex Orica Australia Pty Ltd), an MDI, and 2.7 solid parts of montan wax to form the binder composition.

2. Manufacture of MDF

The binder compositions described above were applied to wood fibres in an MDF process via a blowline. The binder composition was applied at a rate of 28 solid parts of binder to 100 parts of dry wood. The wood fibre was then pressed into MDF panels. The panels of Example 3 were pressed and cured at a temperature of 130°C. The panels of Example 4 were pressed and cured at a temperature of 185°C.

The resultant panel results are as follows:

| Example | Panel Thickness (mm) | Density (kg/m$^3$) | IB (kPa) | 24hr Swell (%) | 70°C Swell (%) | V313 IB (kPa) | V313 Swell (%) |
|---|---|---|---|---|---|---|---|
| 3 | 5 | 779 | 1,900 | 9.6 | 12.5 | 636 | 1.3 |
| 4 | 9 | 938 | 2,020 | 2.3 | 13.1 | 2,020 | 1.1 |

**EXAMPLES 5 and COMPARATIVE EXAMPLE A**

1. Hybrid binder

In Example 5, 100 solid parts of Sylvic DUO A125 (ex Orica Australia Pty Ltd), an MUF resin, was mixed with 17.7 parts of Rubinate DUO B200 (ex Orica Australia Pty Ltd), an MDI, and 1.77 solid parts of montan wax to form the binder composition.

2. Manufacture of MDF

The binder compositions described above were applied to wood fibres in an MDF process via a blowline. The binder composition was applied at a rate of 23.3 solid parts of binder to 100 parts of dry wood. In Comparative Example A QM1329 (ex Orica Australia Pty Ltd), a conventional MUF binder, was applied at a rate of 20.0 solid parts of binder to 100 parts of dry wood. The wood fibre was then pressed into MDF panels.

The resultant panel results are as follows:

| Example | Panel Thickness (mm) | Density (kg/m$^3$) | IB (kPa) | 24hr Swell (%) | 70°C Swell (%) | 100°C Swell (%) | Formaldehyde emission* (mg/100g dry board) |
|---|---|---|---|---|---|---|---|
| A | 5 | 874 | 1,558 | 14 | 34.3 | 54.1 | 17.7 |
| 5 | 5 | 864 | 2,171 | 7.7 | 14.4 | 19.9 | 9.9 |

**EXAMPLES 6 and COMPARATIVE EXAMPLE B**

1. Hybrid binder

   In Example 5, 100 solid parts of Sylvic DUO A133 (ex Orica Australia Pty Ltd), a PF resin, was mixed with 25 parts of Rubinate DUO B200 (ex Orica Australia Pty Ltd), an MDI, montan wax to form the binder composition.

2. Manufacture of MDF

   The binder compositions described above were applied to wood fibres in an MDF process via a blowline. The binder composition was applied at a rate of 13 solid parts of binder to 100 parts of dry wood. In Comparative Example B the binder composition manufactured using DPL 6323 (ex Orica Australia Pty Ltd) was applied at a rate of 13 solid parts of binder to 100 parts of dry wood. The wood fibre was then pressed into MDF panels.

The resultant panel results are as follows:

| Example | Panel Thickness (mm) | Density ($kg/m^3$) | IB (kPa) | 24hr Swell (%) |
|---|---|---|---|---|
| 6 | 16 | 624 | 579 | 8.3 |
| B | 16 | 612 | 316 | 9.9 |

Those skilled in the art will appreciate that the invention described herein is susceptible to variations and modifications other than those specifically described. It is to be understood that the invention includes all such variations and modifications which fall within its spirit and scope. The invention also includes all of the steps, features, compositions and compounds referred to or indicated in this specification, individually or collectively, and any and all combinations of any two or more of said steps or features.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. A binder composition comprising a formaldehyde-based resin and an isocyanate containing compound wherein said formaldehyde-based resin comprises formaldehyde present in a molar ratio of formaldehyde to comonomer in the range of from about 0.2:1 to about 5:1 and wherein the isocyanate containing compound is present in an amount in the range of from about 0.5 to about 50% by weight of the binder composition.

2. A binder composition according to claim 1 wherein the molar ratio of formaldehyde to comonomer is in the range of about 0.3:1 to about 4:1

3. A binder composition according to claim 2 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 3.5:1.

4. A binder composition according to any one of claims 1 to 3 wherein the isocyanate containing compound is a polyisocyanate.

5. A binder composition according to claim 4 wherein the polyisocyanate is selected from the group consisting of toluene diisocyanate and methylene diisocyanate.

6. A binder composition according to any one of claims 1 to 5 wherein the isocyanate containing compound is present in the binder composition in an amount in the range of from 5 to 30% by weight of the binder composition.

7. A binder composition according to claim 6 wherein the isocyanate containing compound is present in the binder composition in an amount in the range of from 10 to 25% by weight of the binder composition.

8. A binder composition according to any one of claims 1 to 7 wherein the formaldehyde-based resin is a urea formaldehyde resin.

9. A binder composition according to claim 8 wherein the molar ratio of formaldehyde

to comonomer is in the range of from about 0.3:1 to about 1.5:1.

10. A binder composition according to claim 9 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 1.1:1.

11. A binder composition according to claim 10 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 0.9:1.

12. A binder composition according to claim 11 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.45:1 to about 0.75:1.

13. A binder composition according to any one of claims 8 to 12 wherein part of the urea is replaced with melamine.

14. A binder composition according to claim 14 wherein the comonomer comprises urea and melamine wherein the melamine is present in the range of from 0.5 to 60 % weight on solids.

15. A binder composition according to any one of claims 1 to 7 wherein the formaldehyde-based resin is a phenol formaldehyde resin.

16. A binder composition according to claim 15 wherein the phenol formaldehyde resin is formulated for acid curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 1:1.

17. A binder composition according to claim 16 wherein the phenol formaldehyde resin is formulated for acid curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 0.9:1.

18. A binder composition according to claim 16 wherein the phenol formaldehyde resin is formulated for acid curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 0.7:1.

19. A binder composition wherein the phenol formaldehyde resins formulated for alkaline curing according to claim 25 wherein in the molar ratio of formaldehyde to comonomer in the range of from about 1.8:1 to about 4:1.

20. A binder composition according to claim 19 wherein the phenol formaldehyde resin is formulated for alkaline curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 2:1 to about 2.5:1.

21. A binder composition according to claim 20 wherein the phenol formaldehyde resin is formulated for alkaline curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 3.3:1 to about 3.8:1.

22. A binder composition according to any one of claim 15 wherein part of the phenol is replaced with urea.

23. A binder composition according to claim 22 wherein the comonomer comprises phenol and urea wherein the urea is present in the range of from 1 to 25 % weight on solids.

24. A binder composition according to any one of claims 1 to 23 wherein the formaldehyde-based resin is blended with a release agent.

25. A binder composition according to claim 24 wherein the release agent is selected from the group consisting of paraffinic and synthetic waxes, polyethylene waxes and polypropylene waxes.

26. A composite panel comprising cellulose-based particles and a binder composition comprising a formaldehyde-based resin and an isocyanate containing compound wherein said formaldehyde-based resin comprises formaldehyde present in a molar ratio of formaldehyde to comonomer in the range of from about 0.2:1 to about 5:1 and wherein the isocyanate containing compound is present in an amount in the range of from about 1 to about 50% by weight of the

binder composition.

27. A composite panel according to claim 26 wherein the molar ratio of formaldehyde to comonomer is in the range of about 0.3:1 to about 4:1

28. A composite panel according to claim 27 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 3.5:1.

29. A composite panel according to any one of claims 26 to 28 wherein the isocyanate containing compound is a polyisocyanate.

30. A composite panel according to claim 29 wherein the polyisocyanate is selected from the group consisting of toluene diisocyanate and methylene diisocyanate.

31. A composite panel according to any one of claims 26 to 30 wherein the isocyanate containing compound is present in the binder composition in an amount in the range of from 5 to 30% by weight of the binder composition.

32. A composite panel according to claim 31 wherein the isocyanate containing compound is present in the binder composition in an amount in the range of from 10 to 25% by weight of the binder composition.

33. A composite panel according to any one of claims 26 to 32 wherein the formaldehyde-based resin is a urea formaldehyde resin.

34. A composite panel according to claim 33 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.3:1 to about 1.5:1.

35. A composite panel according to claim 34 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 1.1:1.

36. A composite panel according to claim 35 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 0.9:1.

37. A composite panel according to claim 36 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.45:1 to about 0.75:1.

38. A composite panel according to any one of claims 33 to 37 wherein part of the urea is replaced with melamine.

39. A composite panel according to claim 38 wherein the comonomer comprises urea and melamine wherein the melamine is present in the range of from 0.5 to 60% weight on solids.

40. A composite panel according to any one of claims 26 to 39 wherein the formaldehyde-based resin is a phenol formaldehyde resin.

41. A composite panel according to claim 40 wherein the phenol formaldehyde resin is formulated for acid curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 1:1.

42. A composite panel according to claim 41 wherein the phenol formaldehyde resin is formulated for acid curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.45:1 to about 0.9:1.

43 . A composite panel according to claim 42 wherein the phenol formaldehyde resin is formulated for acid curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 0.75:1.

44. A composite panel wherein the phenol formaldehyde resins formulated for alkaline curing according to claim 40 wherein in the molar ratio of formaldehyde to comonomer in the range of from about 1.8:1 to about 4:1.

45. A composite panel according to claim 44 wherein the phenol formaldehyde resin is formulated for alkaline curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 2:1 to about 3.5:1.

46. A composite panel according to claim 45 wherein the phenol formaldehyde resin is formulated for alkaline curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 3.3:1 to about 3.8:1.

47. A composite panel according to any one of claims 40 to 46 wherein part of the phenol is replaced with urea.

48. A composite panel according to claim 47 wherein the comonomer comprises phenol and urea wherein the urea is present in the range of from 1 to 25% weight on solids.

49. A composite panel according to any one of claims 26 to 48 wherein the formaldehyde-based resin is blended with a release agent.

50 A composite panel according to claim 49 wherein the release agent is selected from the group consisting of paraffinic and synthetic waxes, polyethylene waxes and polypropylene waxes.

51. A method of manufacturing a composite panel blending cellulose-based particles with a binder composition comprising a formaldehyde-based resin and an isocyanate containing compound wherein said formaldehyde-based resin comprises formaldehyde present in a molar ratio of formaldehyde to comonomer in the range of from about 0.2:1 to about 5:1 and wherein the isocyanate containing compound is present in an amount in the range of from about 1 to about 50% by weight of the binder composition; and curing the binder composition to form a composite board.

52. A method of manufacturing a composite panel according to claim 57 wherein the molar ratio of formaldehyde to comonomer is in the range of about 0.3:1 to about 4:1

53 A method of manufacturing a composite panel according to claim 52 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 3.5:1.

54. A method of manufacturing a composite panel according to any one of claims 51 to

53 wherein the isocyanate containing compound is a polyisocyanate.

55. A method of manufacturing a composite panel according to claim 54 wherein the polyisocyanate is selected from the group consisting of toluene diisocyanate and methylene diisocyanate.

56. A method of manufacturing a composite panel according to any one of claims 51 to 55 wherein the isocyanate containing compound is present in the binder composition in an amount in the range of from 5 to 30% by weight of the binder composition.

57. A method of manufacturing a composite panel according to claim 56 wherein the isocyanate containing compound is present in the binder composition in an amount in the range of from 10 to 25% by weight of the binder composition.

58. A method of manufacturing a composite panel according to any one of claims 51 to 57 wherein the formaldehyde-based resin is a urea formaldehyde resin.

59. A method of manufacturing a composite panel according to claim 58 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.3:1 to about 1.5:1.

60. A method of manufacturing a composite panel according to claim 59 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 1.1:1.

61. A method of manufacturing a composite panel according to claim 60 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 0.9:1.

62. A method of manufacturing a composite panel according to claim 61 wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.45:1 to about 0.75:1.

63. A method of manufacturing a composite panel according to any one of claims 58 to 62 wherein part of the urea is replaced with melamine.

64. A method of manufacturing a composite panel according to claim 63 wherein the comonomer comprises urea and melamine wherein the melamine is present in the range of from 0.5 to 60% weight on solids.

65. A method of manufacturing a composite panel according to any one of claims 51 to 57 wherein the formaldehyde-based resin is a phenol formaldehyde resin.

66. A method of manufacturing a composite panel according to claim 65 wherein the phenol formaldehyde resin is formulated for acid curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 1:1.

67. A method of manufacturing a composite panel according to claim 66 wherein the phenol formaldehyde resin is formulated for acid curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.45:1 to about 0.9:1.

68. A method of manufacturing a composite panel according to claim 67 wherein the phenol formaldehyde resin is formulated for acid curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 0.4:1 to about 0.75:1.

69. A method of manufacturing a composite panel wherein the phenol formaldehyde resins formulated for alkaline curing according to claim 65 wherein in the molar ratio of formaldehyde to comonomer in the range of from about 1.8:1 to about 4:1.

70. A method of manufacturing a composite panel according to claim 69 wherein the phenol formaldehyde resin is formulated for alkaline curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 2:1 to about 3.5:1.

71. A method of manufacturing a composite panel according to claim 70 wherein the phenol formaldehyde resin is formulated for alkaline curing wherein the molar ratio of formaldehyde to comonomer is in the range of from about 2.5:1 to about 3:1.

72. A method of manufacturing a composite panel according to any one of claims 36 to 24 wherein part of the phenol is replaced with urea.

73. A method of manufacturing a composite panel according to claim 72 wherein the comonomer comprises phenol and urea wherein the urea is present in the range of from 1 to 25% based on the molar amount of the comonomer.

74. A method of manufacturing a composite panel according to any one of claims 51 to 73 wherein the formaldehyde-based resin is blended with a release agent.

75. A method of manufacturing a composite panel according to claim 50 wherein the release agent is selected from the group consisting of paraffinic and synthetic waxes, polyethylene waxes and polypropylene waxes.

76. A method of manufacturing a composite panel according to claims 51 to 75 wherein the binder composition is premixed.

77. A method of manufacturing a composite panel according to claims 51 to 75 wherein the formaldehyde-based resin and the isocyanate containing compound are added simultaneously to the cellulose-based particles.

78. A method of manufacturing a composite panel according to claims 51 to 75 wherein the formaldehyde-based resin and the isocyanate containing compound are added separately to the cellulose-based particles.

# INTERNATIONAL SEARCH REPORT

International application No.
PCT/AU 99/00928

| | |
|---|---|
| **A.** | **CLASSIFICATION OF SUBJECT MATTER** |
| Int Cl[6]: | C08G 18/54, 8/28, 12/40, B27N 3/04, 1/02 |

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)
IPC: C08G 18/54, 8/28, 12/40

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
WPAT: IPC as above and urea formaldehyde or phenol formaldehyde and isocyanate: or diisocyanate: or polyisocyanate:

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|---|---|---|
| X | US 5 011 886 A (BUSCHFELD et al) 30 April 1991<br>Whole document | 1-7, 15-20, 24-32, 40-45, 49-57, 65-71, 74-78 |
| X | US 4 268 649 A (JELLINEK et al) 19 May 1981<br>Whole document | 1-7, 15-20, 24-32, 40-45, 49-57, 65-71, 74-78 |
| X | US 4 201 835 A (JELLINEK et al) 6 May 1980<br>Whole document | 1-7, 15-20, 24-32, 40-45, 49-57, 65-71, 74-78 |

[X] Further documents are listed in the continuation of Box C

[X] See patent family annex

* Special categories of cited documents:
- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier application or patent but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed
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- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
- "&" document member of the same patent family

| | |
|---|---|
| Date of the actual completion of the international search<br>23 November 1999 | Date of mailing of the international search report<br>-9 DEC 1999 |
| Name and mailing address of the ISA/AU<br>AUSTRALIAN PATENT OFFICE<br>PO BOX 200<br>WODEN ACT 2606<br>AUSTRALIA<br>Facsimile No.: (02) 6285 3929 | Authorized officer<br><br>GAYE HOROBIN<br>Telephone No.: (02) 6283 2069 |

INTERNATIONAL SEARCH REPORT

International application No.
PCT/AU 99/00928

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|---|---|---|
| X | EP 232 642 A (Societe Chiminique des Charbonnages S.A.) 19 August 1987<br>Whole document | 1-14, 24-38, 49-64, 74-78 |

# INTERNATIONAL SEARCH REPORT
Information on patent family members

| International application No. |
|---|
| PCT/AU 99/00928 |

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

| Patent Document Cited in Search Report | | Patent Family Member | | | | | |
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| US | 5011886 | AT | 41169 | CA | 1265886 | DE | 3346153 |
| | | DK | 6166/84 | EP | 146881 | FI | 845059 |
| | | NO | 845206 | | | | |
| US | 4201835 | AT | 2032/78 | BE | 866019 | CH | 630841 |
| | | DE | 2716971 | DK | 1645/78 | FR | 2387117 |
| | | IT | 1102017 | SE | 7714167 | US | 4268649 |
| | | YU | 595/78 | | | | |
| EP | 232642 | AT | 85061 | DE | 3687652 | ES | 2053453 |
| | | FR | 2592382 | | | | |
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